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Invertebrates of ancient heavy metal spoil and smelting tip sites in southern Jordan: Their distribution and use as bioindicators of metalliferous pollution derived from ancient sources

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The invertebrate populations of stony desert sites contaminated by ancient metalliferous activities in the Nabatean, Roman and Byzantine periods were significantly different from those found in adjacent ancient abandoned field systems. Consequently, it is apparent that such invertebrates potentially have uses as biomonitors of metalliferous pollution derived from ancient anthropogenic activities. This is further explored by an analysis of the heavy metal content of certain of these animals; extensive bioaccumulation of copper and especially lead occurred in organisms inhabiting areas associated with ancient metalliferous enterprises.

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Keywords: Jordan; desert; copper; lead; invertebrates; pollution; biomonitoring; bioaccumulation

Introduction

The important Dana Reserve, which is managed by the Jordanian Royal Society for the Conservation of Nature, is located in southern Jordan. This study area, which is some 70 km to the south-east of the Dead Sea, lies within the Hashemite Kingdom of Jordan. Further detailed information on this region is presented by Barker *et al.* (1997, 1998, 1999) and Pyatt *et al*. (1999, 2000). Pyatt *et al*. (2000) note that this isolated stony desert area is part of the important Wadi Arabah rift valley which represents a continuation of the Red Sea fault. The Dana Reserve is periodically drained by major wadis, including Wadi Dana, Wadi Ghuwayr and Wadi Shegar. These wadis drain limestone, sandstone and basaltic areas (located to the east and north) into Wadi Arabah. Pyatt *et al*. (2000) note that there are rock strata present in the geological sequence which are especially rich in copper, lead, manganese, zinc, iron and tin.

Pyatt *et al*. (1999) noted that from the Bronze Age and thence through the Iron

Age, Nabatean, Roman and Byzantine periods, extensive mining, particularly for copper occurred. The legacy of such extensive ancient metallurgical enterprises remains today as often massive spoil and smelting tips where cations including copper, lead and manganese are present in high concentrations in both the spoil tips and the associated adjacent sediments. Hauptmann *et al*. (1992) indicated that the sedimentary ore deposit at Feinan has been exploited since the seventh millennium BC; they recorded peaks of copper production in the Early Bronze Age, the Iron Age and the Roman period. Pyatt *et al*. (1999) have described how such spoil tips may lose cations by a veriety of processes including sheet and gully erosion, atmospheric erosion, leaching, etc. and this will subsequently lead to pollution/contamination of sites, both adjacent and distant from the spoil and smelting tips. Stretching away from some of the spoil and smelting tip sites are the remains of an extensive field system. This field systems, with its associated irrigation channels, is believed to have been employed for the production of food for the mining and associated military populations inhabiting and exploiting the area in Nabatean, Roman and Byzantine periods. However, it does also incorporate traces of even earlier agricultural systems (Barker et al., 1997, 1998, 1999).

The soils were largely formed from alluvium draining the complex escarpment geology and are composed of clay–sand sized particles eroded from granite, sandstone and limestone parent materials. The vegetation has both a limited diversity and limited biomass per unit area as is not unexpected in such an arid stony desert environment. Information concerning the more important species of this area is presented by Pyatt *et al*. (1999).

Experimental procedure

Although some desert ecosystems are more complex and species-rich than may be initially anticipated, invertebrate population densities are often very low and extremely patchy. Consequently, several studies have used pitfall traps, which can be left in the environment for some time, to produce viable results. Using this technique, Mikhail (1993) was able to estimate soil fauna in a Southern Egyptian wadi, Ayal & Merki (1994)collected over 33 tenebrionid beetle species in the Negev Highlands, and Tigar & Osborne (1999*a,b*) collected 58 taxa of invertebrates near Abu Dhabi.

In the present survey, pitfall traps containing alcohol and water were established during the spring of 1999 and 2000 on spoil tip/smelting sites, and in parts of the field system at varying distances from the more extremely polluted heavy metal processing areas. Replicates were employed and the traps were carefully positioned under stones collected from the site to be investigated. The traps were emptied every 24 h and the invertebrates transferred to the laboratory in sterile sealed containers. The results are derived from a 3/4 day programme of trapping in each case.

The following organisms were further investigated in terms of their bioaccumulation of heavy metals derived from the spoil tip environment:

Cataglyphis sp. (desert ant), *Alopecosa* sp. (wolf spider), *Xisticus* sp. (crab spider), a nymphal and an adult cricket (Orthoptera, Tettigoniidae). These animals were collected from three sites (Table 4) as follows: (1) A 'control' site located some 2 km south-south-east of Khirbet Faynan (a major archaeological feature in close proximity to major spoil tips); (2) a large spoil tip situated 1 km south-west of Khirbet Faynan and (3) the above-mentioned field system. Organisms of similar size and condition were selected; they were thoroughly and carefully washed in sterilized deionized water, dissected and analysed by means of a Perkin-Elmer 1100 Flame Atomic Absorption Spectrophotometer following acid digestion. The results are presented in Table 4 and five replicates were employed in each case.

Results

A total of 27 invertebrate texa were recorded from the seven pitfall sites (Table 1). These included two molluscs (snails), eight arachnids (spiders, mites, ticks and a harvestman), the rest being insect. Due to the paucity of appropriate keys, only ten were identified to genus or species, although sufficient is known about all of the groups to carry out a detailed habitat analysis. (Habitat and distribution information is given in Table 2.)

Table 3 indicates the species richness and diversity data for the sites, and indicates that site 3 (the spoil/smelting area) had by far the greatest range of taxa, the largest numbers, and thus the highest diversity as measured by the Shannon–Weiner index (*H*). In contrast, the spoil tip sites were the most species-poor and least diverse of the areas examined.

A simple similarity analysis of sites 1–7 (% similarity, using the presence/absence of taxa only) indicated the highest similarity between sites 1 and 3 (spoil heap and slag, 286%) and 2 and 7 (spoil heap and old field site, 286%). The lowest similarity

overall was by site 1, since it had no taxa in common (except for two springtails at site 3) with any other site. This analysis could not be taken any further, partly because of this zero similarity and partly because other sites were very species-poor.

Since sites 4–7 were all on the old field system, data from these areas were combined, and an independent *t*-test carried out. This showed that there was a significant difference in the mean number of invertebrates between the smelting tip and field sites (*t* = 07, *p* =004), the second spoil tip and smelting site (*t* = 236,þ^{2} *p* =002) and between the second spoil tip and the field site (*t* = 317, *p* =0003).

A *t*-test does not show how similar the sites are, so a % similarity index was calculated. This showed that the spoil tip site 1 and smelting site 3 were now the most similar (286%), closely followed by the smelting site and the old field system (276%). The second spoil tip (site 2) was much more similar to the field system and

the smelting site than to spoil tip 1. A similarity diagram, using nearest neighbour clustering shows this diagrammatically (Fig. 1).

This form of similarity analysis does not carry any probability, but data were too sparse for correlation, hence the information was reduced again to just three categories: spoil (sites 1 and 2), smelting (site 3) and field (sites 4–7).

A percentage similarity analysis of these combined data indicated that the spoil heaps were 353% similar to the smelting site, and the smelting site 276% similar to

the field site. Spoil and field were 167% similar. At this level, a *t*-test showed no

difference in numbers between spoil and smelting tip (*t* = 026, *p* =08), but the

smelting and field sites did differ significantly (*t* = 207, *p* =004).

A correlation coefficient (Pearson's *r*) showed the smelting and spoil sites to be highly correlated (*r* =+099, *p*o0001), the spoil and field sites to be highly negatively

correlated (*r* = 100, *p*o0001). The old field system and smelting sites were not

correlated (*r* = 04, n/s).

There were too many tied ranks to carry out the more suitable Spearman's rank correlation on these data. **Discussion**

Table 1. *Invertebrates collected by pitfall trapping at Dana Reserve, southern Jordan*

Taxa/description	1 Spoil tip 1	2 Spoil tip 2	3 Spoil smelt	4 Pitfall site 1	5 Pitfall site 2	6 Pitfall site 3	7 Field syst
Mollusca Helicidae *Cernuella virgata*				1			
Mollusca Sphincterochilidae *Albea candidissima*						1	
Aranea Thomisidae *Xisticus* sp. (crab spider)		1					
Aranea Zoariidae *Zoradion* sp. (immature)							1
Aranea Gnaphosidae *Scotophaeus* sp. (immaturre)						1	
Aranea Lycosidae (wolf spiders) *Alopecosa* sp.			1				
Acari Oribatidae (litter mite)	1						
Acari Thrombiciade (earth mite)			1			1	
Acari Metastigmata (tick) (immature)			1				
Opiliones Phalangiidae *Platybunus* sp. (harvestman)			1				
Collembola Entomobryidae sp. 1	1		1				
Collembola Entomobryidae sp. 2	6		3				
Collembola Entomobryidae sp. 3						1	
Thysanura Thermobia sp. (fire brat)					1		
Orthoptera Tettigoniidae (nymphal cricket)						1	
Isoptera Kalotermes sp. (termite worker)			2		1		
Thysanoptera larva (thrip)						1	
Homoptera Auchenorrhyncha Cicadellidae							1
Lepidoptera Tineidae (clothes moth)			1				2
Diptera Phoridae (scuttle fly)				2			
Diptera Sciaridae (fungus gnat)			2				
Hymenoptera Formicidae Formicinae *Cataglyphis* sp.		1	1		4	2	2
Hymenoptera Formicidae Myrmicinae sp. 1		1		1			
Hymenoptera Formicidae Myrmicinae sp. 2					4		
Hymenoptera Formicidae Myrmicinae *Mesor ebeninus*				1	1		
Hymenoptera Formicidae Myrmicinae sp.				5			
Coleoptera Bostricidae (power post beetle)			1				

Table 2. *Habitat preferences of selected species*

Habitat preferences of selected species
Cernuella virgata—Moderately dry and open habitats, calcareous sites, dunes, grasslands, steppe. Mediterranean
Albae candidissima—Grassy hills near coast, preferably bare rocky areas and stony hilltops. North Africa, Syria, Eastern Mediterranean (Pfleger, 1998)
Xisticus sp.—dunes and healthland
Zoradion sp.—desert wolf spider
Scotophaeus sp.—dry, warm habitats, indoors farther north
Alopecosa sp.—grassland and heath
Oribatidae/Thrombicidae—moist environments, decomposing organic matter, may feed on termites (Goodall & Perry, 1979)
Platybunus—in herbage and detritus
Thermobia—Southern Europe, open warm habitats. Detritivore (Zahradnik, 1999)
Field cricket—dry, warm places, underground usually. Eats almost anything. North Africa (Zahradnik, 1999)
Kalotermes—dry wood termites, often in buildings in arid regions (Cloudsley-Thompson, 1977)
Tineidae—larvae in woollen fabrics and dead animals (sheep/goats) in southern areas. Clothes moths.
Phoridae—larvae in dry carrion
Sciaridae—larvae in soil and fungi
Cataglyphis—desert ant. Mediterranean, North Africa, near east (North, 1996)
Myrmicinae 1–4—On Red Sea hills at 40°C (Cloudsley-Thompson, 1977)
Bostricidae—in decaying timber. Middle East, Asia Minor, Central and Southern Europe (Harde, 1998).

Table 3. *Invertebrate abundance and diversity from pitfall analysis*

Category	Spoil 1	Spoil 2	Smelt 3	Field system 4	Field system 5	Field system 6	Field system 7
Taxa	3	3	11	5	5	7	4
Individuals	8	3	15	10	11	8	6
Shannon–Weiner *H*	0736	1100	2300	136	139	191	133

A number of factors control the abundance of invertebrates in arid areas. The chief amongst these are temperature range (affected by the amount of shelter), presence of live and dead organic matter, and especially the availability of water (Noy-Meir, 1985). A more complex site with shelter, food and moisture is likely to be more species-rich than an exposed, dry area, and this is clearly true of the smelting site 3 which contains a wide range of invertebrates, including decomposers and detritivores, as well as desert-adapted animals. The site was on the margin of the smelting tip site and was selected due to its limited smelting remains and possibly limited earlier activity. The site was associated with areas which maintained water during wetter periods of the year, a mixed topography and the presence of various large stones which might conceivably provide microhabitats. At the other, more exposed sites, it is arid habitat species that predominate, for example spoil tip site 2, whose taxa are distinct from all other sites examined. (This site has been described by Pyatt *et al*., 1999.) Spoil tip site 1, however, is quite similar to the smelting site, although again very poor in species. This site is located approximately 1 km south-west of Khirbet Faynan, the major archaeological feature in the area.

Although the old field system sites were grouped together, it is clear that they have low invertebrate similarity, stressing the importance of microhabitats. Pitfall site 4 was located *ca.* 2 km south-south-east of Khirbet Feynan; it has less pollution which is brought in by agencies such as atmospheric erosion. Pitfall site 5 overlooks Zureiq el Mirad and is part of the old abandoned field system. There is no obvious evidence of the remnants of spoil or smelting materials. Pitfall site 6, also in the abandoned field system but low in the valley, only shares the widely distributed ant *Cataglyphis* with the other field sites, and Pitfall site 7 is close to a smelting site, but has little similarity to the spoil smelting site.

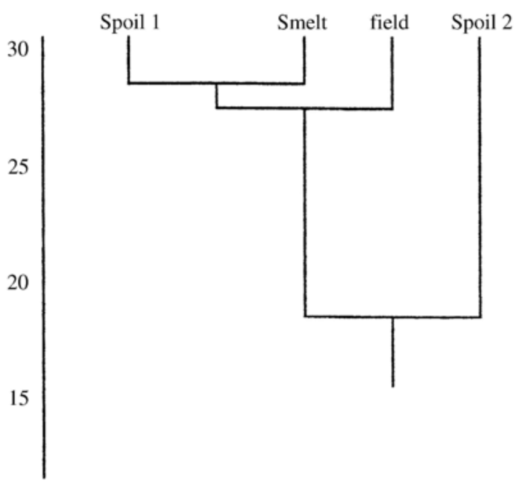

Figure 1. % similarity analysis of invertebrates present at different site types.

Since each of the sites examined had good ground cover, composed of large stones and rocks, differences in species-richness are likely to be caused by food availability, which originates from a number of sources and/or environmental toxicity. Windblown vegetation fragments can form an important part of the food chain in the absence of live plant material (Cloudsley-Thompson, 1977), which often is little utilized by herbivores, only 2–10% being consumed. The rest is removed by invertebrate detritivores such as termites, ants and woodlice, plus weathering (Noy-Meir, 1985). A second source of food is seeds, which are often collected by ants (Goodall & Perry, 1979), although some ants such as *Cataglyphis* species are predatory (North, 1996). Either of these processes could result in the concentration of heavy metals in the detritivores, although there is both positive and negative evidence for this (Mackay *et al*., 1997).

The two other processes which must be considered are the presence of animal dung, fresh or Cold, and dead animal material. Both of these may persist in an arid environment for many years, despite the activities of dung beetles, and dry carrion insects such as scuttle flies (Phoridae), dermestid beetles and moths (Tineidae). This could help to explain why the old field system was generally more diverse than the spoil heaps, although similar to them. Indeed the pattern is complex; dung is derived in this area particularly from both goats and sheep; however, in addition, part of the field system will have an enhanced organic burden as a consequence of earlier activities in terms of the growing of melons.

With regard to the individual animal groups, five taxa (or 19%) were herbivorous, 11 detritivores (41%) and six predatory (22%) There were five taxa of social insect. This contrasts with Tigar & Osborne (1999*b*),whose results included only 4% of herbivores and fewer detritivores (28%). However, 17 species (29%) were predatory, and 36% of the texa consisted of social insects (Hymenoptera, Formicidae), both of which were more frequent at full moon.

The similarity analysis of grouped sites shows that the smelting and spoil tip sites were positively correlated, and the spoil and field sites negatively correlated. However, due to the low numbers of individuals caught, it is not clear at this stage whether similarity and a reduction in diversity as a result of pollution and past activity can be separated from microhabitat differences.

Pitfall trap captures are influenced by the intensity of moonlight, by animals already caught, which attract predators and detritivores, and they can provide only a part of the picture. For example, over 90,000 invertebrates were caught by light trapping in the sandy deserts at Abu Dhabi over a 2 year period (Tigar & Osborne, 1999*a*). However, the present results stress the variability of sites with a similar history as well as indicating that diversity is clearly reduced at the spoil tip sites and hence there exists a potential to utilize such invertebrates in biomonitoring programmes. This is now further explored.

The results from the determination of the concentrations of copper and lead in selected invertebrates, from three sites, are presented in Table 4. It has previously been reported (Pyatt *et al*., 1999, 2000; Pyatt & Grattan, 2001) that both modern vegetation and ancient human skeletal materials, obtained from the study area, contain enhanced concentrations of copper and lead. Examining values in desert invertebrates provides the opportunity to assess processes at a different trophic level as the organisms employed are all essentially herbivores.

Whilst lead is toxic to organisms, copper is essential, in appropriate concentrations, to invertebrates and along with other elements is found in biochromes (Rockstein, 1987).These appear as metalloproteins and play an important role in food synthesis.

A number of points emerge from the results. The lead bioaccumulated by the desert invertebrates is in higher concentration than the copper values and the least bioaccumulation occurred in the legs (essentially composed of muscles and chitinous exoskeleton). The values in the head portions and abdomen portions (both with

Table 4. *Copper and lead concentration of organisms collected from three sites*

	Site 1						Site 2						Site 3					
	Leg		Abdomen		Head		Leg		Abdomen		Head		Leg		Abdomen		Head	
	Cu	Pb	Cu	Pb	Cu	Pb	Cu	Pb	Cu	Pb	Cu	Pb	Cu	Pb	Cu	Pb	Cu	Pb
Hymenoptera Formicidae Formicinae *Cataglyphis* sp. (desert ant)	0·08	2·56	0·07	3·71	0·23	5·1	1·13	2·7	1·94	8·5	2·73	11·3	0·1	2·6	0·05	4·11	0·25	5·72
Aranea Lycosidae *Alopecosa* sp. (wolf spider)	0·06	3·56	0·17	16·9	0·57	9·9	1·6	3·4	2·28	31·3	8·18	21·4	0·08	3·6	0·19	17·32	0·49	8·52
Aranea Thomisidae *Xisticus* sp. (Crab spider)	0·58	4·2	0·67	11·5	0·31	9·91	1·66	4·20	2·28	21·6	8·20	19·72	0·61	4·3	0·73	12·22	0·32	10·02
Orthoptera Tettigoniidae (nymphal *cricket*)	0·82	1·49	1·46	2·33	1·56	2·75	1·85	2·49	2·49	22·83	4·82	21·61	0·91	2·1	1·50	3·92	1·57	4·03
Adult cricket	1·57	1·61	2·85	9·92	2·04	10·01	2·32	4·61	3·94	37·46	9·06	33·01	1·61	2·8	2·61	4·63	2·13	3·82

The results are presented in ppm and are the mean of five replicates in each case.
N.B. Sites for metalliferous work are as follows: *Control Site* 1: 'Control' site located 2 km SSE of Khirbet Faynan; *Metalliferous Site* 2: Spoil tip 1 km SW of Khirbet Faynan; *Field site* 3: Field system.

associated gut tissues) were higherFthese tissues being exposed to heavy metals ingested with the food (Pyatt *et al*., 1999, 2000). In support, Newman & McIntosh (1991) noted that the most important route for metal assimilation in terrestrial invertebrates is via the digestive tract.

The data on the nymphal and adult crickets reflect the fact that bioaccumulation processes, in this animal, become further enhanced with age.

From Table 4, it is also evident that values in site 2 are enhanced as compared to sites 1 and 3; i.e. species inhabiting desert areas in close proximity to metalliferous spoil tips are particularly prone to bioaccumulate cations such as copper and lead. The enhancement may be as great as 30-fold, eg. copper in the abdomen of *Cataglyphis* in polluted site 2 as compared to 'control' site 1. Such areas of desert are prone to severe abiotic effects, and Baranowska (1995) noted that drought and low humidity facilitate intense lead uptake and transport to aerial portions of plants; these are hence readily available to many herbivores.

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